

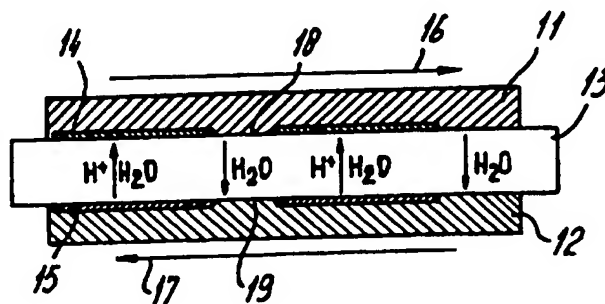
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## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification <sup>6</sup> :</b> <b>H01M 8/04, 8/10</b>	<b>A1</b>	<b>(11) International Publication Number:</b> <b>WO 96/24958</b> <b>(43) International Publication Date:</b> 15 August 1996 (15.08.96)
<b>(21) International Application Number:</b> PCT/NL96/00063 <b>(22) International Filing Date:</b> 9 February 1996 (09.02.96) <b>(30) Priority Data:</b> 9500253 10 February 1995 (10.02.95) NL <b>(71) Applicant (for all designated States except US):</b> STICHTING ENERGIEONDERZOEK CENTRUM [NL/NL]; Westduinweg 3, NL-1755 LE Petten (NL). <b>(72) Inventor; and</b> <b>(75) Inventor/Applicant (for US only):</b> MALLANT, Ronald, Karel, Antoine, Maria [NL/NL]; Koggewaard 83, NL-1824 GP Alkmaar (NL). <b>(74) Agent:</b> DE BRUIJN, Leendert, C.; Nederlandsch Octrooibureau, Scheveningseweg 82, P.O. Box 29720, NL-2502 LS The Hague (NL).		<b>(81) Designated States:</b> AL, AM, AT, AU, AZ, BB, BG, BR, BY, CA, CH, CN, CZ, DE, DK, EE, ES, FI, GB, GE, HU, IS, JP, KE, KG, KP, KR, KZ, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, TJ, TM, TR, TT, UA, UG, US, UZ, VN, ARIPO patent (KE, LS, MW, SD, SZ, UG), Eurasian patent (AZ, BY, KG, KZ, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i> <i>In English translation (filed in Dutch).</i>

**(54) Title:** SOLID POLYMER FUEL CELL COMPRISING HUMIDITY-EXCHANGING AREAS**(57) Abstract**

For the purpose of humidifying anode gases (17) with the aid of moisture present or in cathode gases (16) taken up it is proposed that the gases, in the cell or in the direct vicinity thereof, be caused to enter a humidity-exchanging relationship. In the case of the method being implemented in the cell, the anode (12) and the cathode (11) are designated as strips provided with catalyst (14, 15) and the areas (18, 19) inbetween as portions which are not provided with catalyst. These portions not provided with catalyst, which are non-active or less active, provide for optimal water permeability.



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Solid polymer fuel cell comprising  
humidity-exchanging areas

The present invention relates to a solid polymer fuel cell, comprising an anode, electrolyte/diaphragm and cathode, and  
5 a supply and exhaust of gases on both the anode and the cathode side, the anode comprising areas provided with catalyst for carrying out the electrochemical reaction and, adjacent thereto, areas not provided with catalyst for absorbing moisture via electrolyte/diaphragm.

10 Such a device is known from US Patent No. 4,973,530.

It is necessary for anode gases, in particular, to be humidified, in order to keep the polymer/electrolyte sufficiently moist, so that optimal ion conductivity in the cell can occur and the latter thus has an adequate output. This is because, during  
15 operation of a solid polymer fuel cell, the transport of  $H^+$  is accompanied, simultaneously, with the transport of water in the same direction. Although the phenomenon of water molecules being carried along by protons is compensated for, to some extent, by a diffusion of water molecules back to the anode, depletion of water  
20 takes place at the anode side of the cell with a corresponding poor conductivity for protons and a decrease in the output and performance of the cell.

The abovementioned US Patent No. 4,973,530 proposes to cause the gases to cover a serpentine trajectory across the  
25 electrode in question. The electrode which is provided with catalyst is bordered by an area which is not provided with electrolyte material, but where a water-exchanging diaphragm is present. Since the serpentine always covers a portion of the area where the water-exchanging diaphragm is present, it is possible  
30 for water to be taken up at the anode side. The converse process takes place at the cathode side, water being given off via a water-exchanging membrane and a water-absorbing medium.

It will be understood that at the anode side, based on this supply of water in the direction of the cell up to the point  
35 where the serpentine again reaches the water supply, the water concentration will decrease. Thus, a kind of sawtooth-like profile of the moisture concentration in the cell will be obtained. As a result, performance and output will have a corresponding profile.

The drawback of such cells is the presence of serpentine

which require expensive separate processing steps.

In the embodiment according to said US Patent, four different mass flows are present: the anode gas, the cathode gas, the medium (water) for humidifying the anode gas, and the medium  
5 for removing the water from the cathode gas. These mass flows need to be kept separate from one another in a reliable manner.

Moreover, the medium which has removed the water from the cathode gas stream should be stripped of water, before said water can be passed through the humidifying sections of the anode  
10 stream. This requires further expensive provisions.

The object of the present invention is to provide a fuel cell wherein such a serpentine supply or exhaust of gases is unnecessary and which can therefore be accomplished in a simple manner. Moreover, the depletion in terms of water vapour should be  
15 compensated for at each point of the electrode and not just at one point.

This object is achieved, in an above-described solid polymer fuel cell, in that the areas provided with catalyst comprise strips which are bounded, at least on their long sides,  
20 by areas not provided with catalyst, which areas without catalyst have a minimum width of 1 mm.

By virtue of the portions provided with catalyst being designed as strips which are bordered by non-active or less active portions as a result of the absence of catalyst, provision is  
25 effectively made, at each point of the active portion of the electrode, for compensation of the depletion in moisture by water molecules being carried along together with  $H^+$ .

It should be noted that the Japanese Patent Abstract No. 58-126675 discloses the use of a platinum mesh disposed on the  
30 diaphragm. Such a mesh is used to increase vibration resistance. Although this publication does not give any dimensions, it can be assumed, presuming that the thickness of the membrane is between 100 and 200  $\mu m$ , that the spacing of the platinum wires is in the order of 50  $\mu m$ , in contrast to the present invention where the  
35 spacing between the areas provided with catalyst is at least 1 mm. If the spacing is as small as shown in the "Abstract" of the Japan se Application No. 58-126675, the effect of certain areas not being active is not produced, and that entire portion of the diaphragm which is covered with platinum gauze, should be regarded

as one active area for the electrochemical reaction.

The European Application No. 0 654 837 A1, published after the priority date of the present application, proposes a saving in catalyst. In this case, the supply of gases takes place via corrugations, and where the corrugations touch the electrodes, the gases will be less active. Precisely at those points, less catalyst is applied. This, however, requires the accurate positioning, on top of one another, of the areas which are not provided with catalyst and of the corrugation. Moreover, in such areas it will also not be possible to move water back to the anode side, because of the obstructing portion of the corrugation.

The above-described design is of particular importance at the anode side because it is there that water depletion takes place.

However, it has proved to be advantageous for the cathode side to be designed in the same manner.

In order to optimize, as far as possible, the effect of water being moved back, the gas supply at the anode and the cathode, respectively, is preferably positioned so as not to be parallel to the strips in question. More in particular, it is perpendicular to the strips. The strips may be straight but may likewise be of any shape known in the prior art, such as a zigzag shape.

The invention also relates to a method for fabricating an above-described anode and/or cathode. In the process, there are provided a support not provided with catalyst and a slurry comprising carbon and catalyst, the slurry being applied to the support or to the polymer/electrolyte, in the desired pattern, by printing.

Of course, it is also possible to use other methods to apply the pattern in question to a support or to the polymer/electrolyte. By means of the invention the advantage is achieved that the cathode gases and anode gases enter a direct humidity-exchanging relationship, without use being made of water-separating facilities which are customarily incorporated in the cathode exhaust gas stream, without means for transferring said water to chambers which enter into a humidity-exchanging relationship with the anode gases, and without the need for compartments incorporated in the cell or in the immediate vicinity thereof and

intended to enable humidity exchange with cathode gas or anode gas.

Thus, it is possible for the catalyst (slurry) to be applied to the support or directly to the polymer, the support then being positioned against the polymer electrolyte provided with catalyst.

The invention is explained below in more detail with reference to the illustrative embodiments shown in the drawing, in which

Fig. 1 shows diagrammatically, in section, an SPFC cell, humidity exchange between cathode and anode gases taking place at the edge of the cell,

Fig. 2 shows, on a reduced scale, the entire cell according to Fig. 1 in top view;

Fig. 3 shows a second embodiment of the invention in section, exchange of humidity taking place in the cell;

Fig. 4 shows a top view, on a reduced scale, of the cell according to Fig. 3; and

Fig. 5 shows a top view of a variation of the embodiment according to Figs. 3 and 4.

Fig. 1 schematically depicts a detail of a solid polymer fuel cell. Only those parts are shown, which are important for understanding the present invention.

This cell is composed of a cathode 1 and an anode 2 between which a polymer diaphragm/electrolyte 3 is inserted. The cathode and anode are provided with catalyst, respectively indicated by 4 and 5. Arrow 6 indicates the motion of the oxygen-containing gas stream, whereas arrow 7 shows the movement of the fuel gas. It should be understood that both gases are passed over the electrodes via some sort of duct system. Likewise it should be understood that the indicated direction of movement of anode gases and cathode gases should be seen as preferred directions for obtaining an optimum result, but that other directions likewise fall within the scope of the invention described herein. Moreover, the cell may be a stacked cell.

Fig. 1 shows that there is no catalyst near the edge of the fuel cell. There, the anode and cathode are non-active with respect to effecting proton transport and are particularly porous. The cathode gas, whose oxygen has been consumed in its entirety or

in part contains, near the edge, a very high proportion of water. Owing to the anode gas, represented by arrow 7, being relatively dry, diffusion of water through electrolyte 3 will take place. In Fig. 2, the embodiment according to Fig. 1 is shown in its entirety in a view reduced in size. This shows that the left-hand edge area is intended for the diffusion, whereas the right-hand portion comprises the fuel cell proper. In the left-hand edge portion, water will move from cathode 1 to anode 2 and in the right-hand edge portion in exactly the opposite way.

Fig. 3 shows a further alternative of the invention. In this figure, the movement of water is likewise represented schematically.

The cathode is indicated herein by 11, the anode by 12, the electrolyte by 13, the catalysts by 14 and 15, whereas the arrow for oxygen-containing gas is indicated by 16 and the arrow for fuel gas by 17. It is clear that this cell contains areas (indicated by 18 and 19) where there is no catalyst, i.e. which are non-active or less active.

This figure shows that in the areas which are active and provided with catalyst, transport of water takes place from the anode 12 to the cathode 11. This gives rise to humidification of the cathode gas and water depletion of the anode gas. In the non-active portions precisely the opposite transport takes place. Because no reaction takes place there, transport of water will be determined exclusively by a diffusion mechanism and will not be prevented by the flow of protons.

By situating such non-active areas in the cell it is possible to provide, even in situ enrichment with water. As Fig. 3 shows, continuous humidification of the anode gases takes place and it is not necessary to add separate installations for removing the humidity from the cathode gas.

It should be understood that the figures are not drawn to scale. The thickness of the diaphragm 3 and 13, respectively, is usually in the range between 25 and 200  $\mu\text{m}$ . The thickness of the electrodes is approximately 00 [sic]-400  $\mu\text{m}$ .

Fig. 4 shows a top view of the cathode on a reduced scale. This shows that a series of areas which are less active alternates with a series of areas or strips which are active. The width of an active strip can be between 6 and 10 mm, whereas the

width of the portion not provided with catalyst is greater than 1 mm and is preferably 2 to 3 mm. Generally it can be assumed that the ratio not covered with catalyst/covered is  $1/5 - 1/3$ . Thus the distance which the water has to cover can be made as short as possible. In order, moreover, to make the design as robust as possible and achieve further optimization, the design according to Fig. 5 can be applied, in which 20 indicates the active areas.

It will be understood that the design shown in Figs. 4 and 5 obviously, and in the first instance, also applies to the anode side.

Although it has been stated, in the above, that the intended transport of humidity from cathode back to anode can be achieved by simply omitting the catalyst, this can be realised with any method known in the prior art. Thus it is possible for the electrode, in the areas in question, to be removed in its entirety.

The design shown with reference to Figs. 1-5 can be applied to each cell or a series of cells.

The above-described pattern of strips provided with catalyst and strips not provided with catalyst can be applied to the electrode in any manner known in the prior art. Preferably, printing is employed, since the slurry used for the catalyst can be adapted in a simple manner so as to have rheological properties comparable with printing ink. Thus, printing can be effected in a simple, accurate manner. This printing can take place with any method known in the prior art, such as screen printing, rotary printing and jet printing.

It was found that if, with respect to the active area, the non-active area having a surface area of 20% of the active area is used, sufficient recovery of water can be obtained to obviate additional installations. If required, separate means such as cavities may be present to collect the water and to redistribute it. Moreover, it is possible for the active and non-active areas to be completely separate from one another and to be situated at some distance from one another. The feature essential for the invention, which is that the cathode gases serve as a humidity (water) carrier, is maintained in such an embodiment.

These and other variations are deemed to be within the scope of the accompanying claims.



Claims

1. Solid polymer fuel cell, comprising an anode (12), electrolyte/diaphragm (13) and cathode (11), and a supply and exhaust of gases on both the anode and the cathode side, the anode  
5 comprising areas provided with catalyst (15) for carrying out the electrochemical reaction and, adjacent thereto, areas (19) not provided with catalyst for absorbing moisture via electrolyte/diaphragm, characterized in that said areas provided with catalyst comprise strips which are bounded, at least on their long  
10 sides, by areas not provided with catalyst, which areas without catalyst have a minimum width of 1 mm.
2. Solid polymer fuel cell according to Claim 1, the cathode comprising areas provided with catalyst for carrying out the electrochemical reaction and, adjacent thereto, areas not provided  
15 with catalyst for giving off moisture via the electrolyte/diaphragm, wherein said areas provided with catalyst comprise strips which are bounded, at least on their long sides, by areas not provided with catalyst, which areas without catalyst have a minimum width of 1 mm.
- 20 3. Solid polymer fuel cell (SPFC), wherein the gas supply takes place at the anode and/or cathode at an angle with said strips.
4. Solid polymer fuel cell according to Claim 3, wherein the gas supply takes place at the anode and/or cathode essentially  
25 perpendicular to said strips.
5. Solid polymer fuel cell, wherein said strips are designed in a zigzag shape.
6. Solid polymer fuel cell according to any one of the preceding claims, wherein the catalyst comprises platinum.
- 30 7. Method for fabricating an anode/cathode according to any one of the preceding claims, comprising the provision of a support not provided with catalyst and of a slurry comprising carbon and catalyst, wherein the suspension is applied to the support, in the desired pattern, by printing.

1/1

fig-1

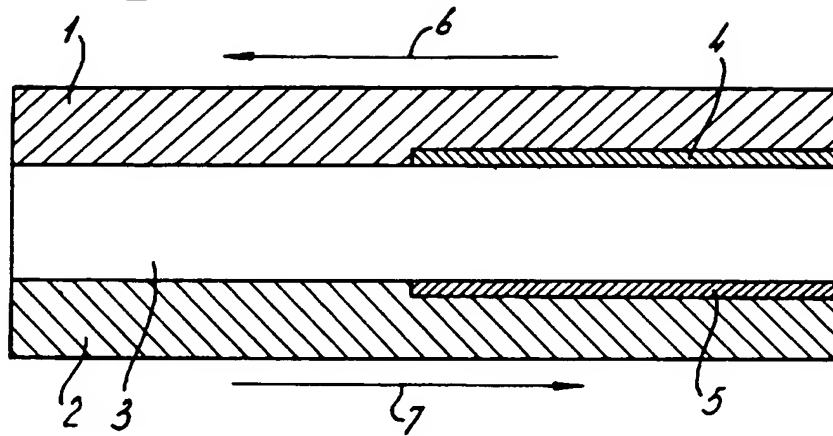


fig-2

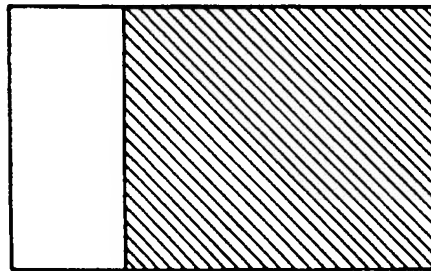


fig-3

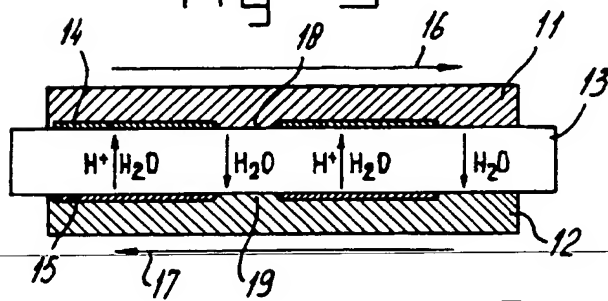


fig-4

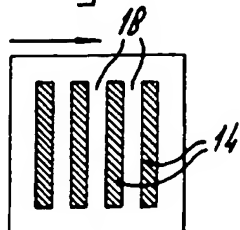
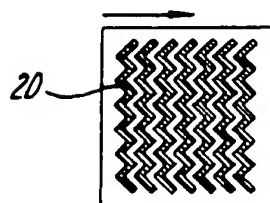


fig-5



# INTERNATIONAL SEARCH REPORT

Internat Application No  
PCT/NL 96/00063

A. CLASSIFICATION OF SUBJECT MATTER  
IPC 6 H01M8/04 H01M8/10

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP,A,0 275 465 (DOW CHEMICAL CO) 27 July 1988 see column 16, line 1 - line 9; claims 1,2,4,15; figure 3 see column 6, line 4 - line 12 see column 8, line 43 - line 52 ---	1,2,6
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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

24 May 1996

Date of mailing of the international search report

31.05.96

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# INTERNATIONAL SEARCH REPORT

Internat. Application No.  
PCT/NL 96/00063

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Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	AT,B,389 020 (SCHUETZ PETER DIPL ING DR) 10 October 1989 see page 2, last paragraph - page 3, paragraph 1 see page 3, paragraph 3 see page 5, line 15 - line 36 ---	1
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Information on patent family members

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European Patent  
Office

**SUPPLEMENTARY  
EUROPEAN SEARCH REPORT**

Application Number  
EP 99 95 1363

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
P,X	WO 99 05741 A (DUBOSE RONALD A ;EMPRISE CORP (US)) 4 February 1999 (1999-02-04) * page 3, line 8-25 * * page 9, line 25 - page 10, line 5 * * figures *	1,6,8,17	H01M8/04 H01M8/06 H01M8/10
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The supplementary search report has been based on the last set of claims valid and available at the start of the search.			
Place of search <b>MUNICH</b>		Date of completion of the search <b>2 August 2001</b>	Examiner <b>Engl, H</b>
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document			

3

EPO FORM 1503 03.82 (P04C04)

**ANNEX TO THE EUROPEAN SEARCH REPORT  
ON EUROPEAN PATENT APPLICATION NO.**

EP 99 95 1363

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report.  
The members are as contained in the European Patent Office EDP file on  
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02-08-2001

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